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(54) Title: SURFACE-ACTIVE FORMULATIONS

(57) Abstract

The invention relates to surface-active soap formulations, comprising (a) 0.01 to 0.2 % by weight of a microbicidal active substance, (b) 0.1 to 7.5 % by weight of one or more than one hydrotropic agent, (c) 0 to 2 % by weight of one or more than one synthetic surface-active substance or of a soap or of combinations of the cited substances and/or of a salt of a saturated and/or unsaturated C_8-C_{22}fatty acid, (d) 0 to 10 % by weight of a dihydric alcohol, (e) 0 to 70 % by weight of a monohydric alcohol, and (f) mains water or deionised water to make up 100 %. The formulation is used for the disinfection and cleansing of the human skin and hands and of hard objects.

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Surface-active formulations

The present invention relates to surface-active formulations as well as to their use for the disinfection and cleansing of the human skin and hands and of hard objects.

Special demands of hygiene are not only in hospitals, restaurants or in the food sector, but also in the private sector, as for example in private households or when travelling whereby the demands made upon the disinfectants employed are undoubtedly less stringent in these sectors than in the first-mentioned group. Nonetheless these disinfectants should act within as short a time as possible, and they should preferably be worth the money, have good skin compatibility and be ecologically safe.

Surprisingly, it has now been found that a formulation comprising an extremely low concentration of a microbicidal active substance as well as further components, typically surface-active components, has these properties.

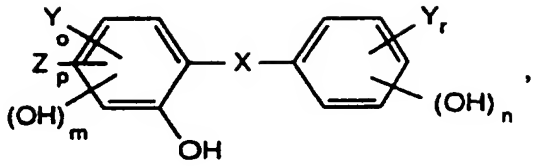
The surface-active formulations comprise

(a) 0.01 to 0.2% by weight of a microbicidal active substance,
(b) 0.1 to 7.5% by weight of one or more than one hydrotropic agent,
(c) 0 to 2% by weight of one or more than one synthetic surface-active substance or of a soap or of combinations of the cited substances and/or of a salt of a saturated and/or unsaturated C_8-C_{22}fatty acid,
(d) 0 to 10% by weight of a dihydric alcohol,
(e) 0 to 70% by weight of a monohydric alcohol, and
(f) mains water or deionised water to make up 100%.

Soap formulations will be understood as meaning aqueous soap solutions which may be obtained as soap or so-called syndet solutions (synthetic detergents).

The antimicrobial activity of the novel formulation reaches gram-positive and gram-negative bacteria as well as yeasts, dermatophytes and the like.

Suitable components (a) are preferably 2-hydroxydiphenyl ethers, 2-hydroxydiphenyl methanes and 2-hydroxydiphenyl thioethers of the general formula

(1) 

wherein

X is oxygen, sulfur or $-CH_2-$,

Y is chloro or bromo,

Z is SO_2H, NO_2 or C_1-C_4alkyl,

r is 0 to 3,

o is 0 to 3,

p is 0 or 1,

m is 0 or 1, and

n is 0 or 1.

Of particular interest are compounds of formula (1), wherein

X is oxygen, sulfur or $-CH_2-$, and

Y is chloro or bromo,

m is 0,

n is 0 or 1,

o is 1 or 2,

r 1 is or 2, and

p is 0.

Of very particular interest are compounds of formula (1), wherein

X is oxygen, and

Y is chloro,

m is 0,

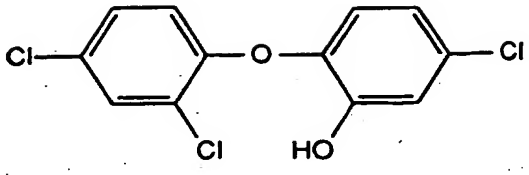
n is 0,

o is 1,

r is 2, and

p is 0.

The compound of formula

(2) 

is very particularly preferred here.

Component (a) in the novel formulation is preferably used in amounts of 0.02 to 0.2% by weight.

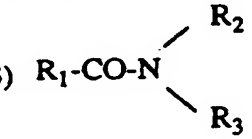
The following compounds are suitable for use as component (b):

(b_1): sulfonates, preferably the salts thereof of terpenoids, or mono- or binuclear aromatic compounds, typically sulfonates of camphor, toluene, xylene, cumene or naphthene;

(b_2): saturated or unsaturated C_3-C_{12}di- or polycarboxylic acids, typically malonic, succinic, glutaric, adipic, pimelic, suberic, azelaic and sebacic acid, undecanedicarboxylic acid and dodecanedicarboxylic acid, fumaric, maleic, tartaric and malic acid as well as citric and aconitic acid;

(b_3): - aliphatic saturated or unsaturated C_1-C_{11}monocarboxylic acids, typically acetic, propionic, hexanoic, capric or undecylenoic acid;
- saturated or unsaturated C_3-C_{12}di- or polycarboxylic acids, typically malonic, succinic, glutaric, adipic, pimelic, suberic, azelaic and sebacic acid, undecanecarboxylic and dodecanedicarboxylic acid, fumaric, maleic, tartaric and malic acid as well as citric and aconitic acid;
- aminocarboxylic acids, typically ethylenediaminetetracetic acid, hydroxyethylethylenediaminetetracetic acid and nitrilotriacetic acid;
- cycloaliphatic carboxylic acids such as camphoric acid;
- aromatic carboxylic acids, typically benzyl, phenylacetic, phenoxyacetic and cinnamic acid, 2-, 3- and 4-hydroxybenzoic acid, anilinic acid as well as o-, m- and p-chlorophenylacetic acid and o-, m- and p-chlorophenoxyacetic acid;
- alkali metal salts and amine salts of inorganic acids, typically the sodium or potassium salts and amine($R_1R_2R_3$) salts of hydrochloric, sulfuric, phosphoric, C_1-C_{10}alkylphosphoric acid and boric acid, in which amine salts R_1, R_2 and R_3 have the meaning indicated above;
- isethionic acid;
- tannic acid;

- acid amides of formula

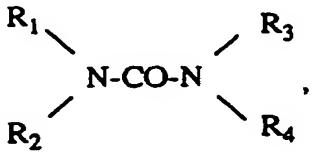
(3)  ,

wherein

R_1 is hydrogen or C_1-C_{12}alkyl, and

R_2 and R_3 are each independently of the other hydrogen, C_1-C_{12}alkyl, C_2-C_{12}alkenyl, C_1-C_{12}hydroxyalkenyl, C_2-C_{12}hydroxyalkyl, or a polyglycol ether chain containing 1 to 30 $-CH_2-CH_2-O-$ or $-CHY_1-CHY_2-O-$ groups, wherein one of the radicals of Y_1 or Y_2 is hydrogen and the other is methyl, e.g. N-methylacetamide;

- urea derivatives of formula

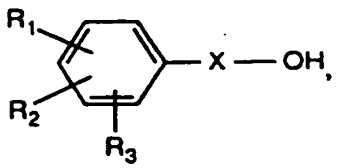
(4)  ,

wherein

R_1, R_2, R_3 and R_4 are each independently of one another hydrogen, C_1-C_8alkyl, C_2-C_8alkenyl, C_1-C_8hydroxyalkyl or C_2-C_8hydroxyalkenyl;

- monohydric C_4-C_{18}aliphatic and monocyclic alcohols, typically C_2-C_{18}alkanols, C_2-C_{18}alkenols and terpene alcohols e.g. ethanol, propanol, isopropanol, hexanol, cis-3-hexene-1-ol, trans-2-hexene-1-ol, 1-octen-3-ol, heptanol, octanol, trans-2-cis-6-nonadien-1-ol, decanol, linalol, geraniol, dihydroterpineol, myrcenol, nopol and terpineol;

- aromatic alcohols of formula

(5)  ,

wherein

X is $-(CH_2)_{1-6}$, $-CH=CH-CH_2-$, or $-O-(CH_2)_{2-6}$, and

R_1, R_2 and R_3 are each independently of one another hydrogen, hydroxy, halogen

or C_1-C_6alkoxy, typically benzyl alcohol, 2,4-dichlorobenzyl alcohol, phenoxyethanol, 1-phenoxy-2-propanol (phenoxyisopropanol) and cinnamyl alcohol;

- polyhydric alcohols and polyhydric alkoxylated, preferably ethoxylated and/or propoxylated alcohols as well as the ethers and esters thereof of the general formula

(6) R_1-O-X-O-R_2,

wherein

R_1 and R_2 are each independently of the other hydrogen, C_1-C_{12}alkyl, C_2-C_{12}alkenyl, C_1-C_8alkanoyl, C_3-C_{18}alkenoyl,

R_3-(OCH-CH_2)$_{1\text{-}50}$, wherein
|
R_4

R_3 is hydrogen, C_1-C_{12}alkyl or C_2-C_{12}alkenyl, and

R_4 is hydrogen or -CH_3, and

X is C_2-C_{10}alkylene or C_2-C_{10}alkenylene, -$(CH_2CH_2O)_{1\text{-}50}$ CH_2-CH_2- or

-(CH_2-CH-O)$_{1\text{-}50}$ CH_2-CH- .
| |
CH_3 CH_3

All organic acids mentioned under (b) may also be obtained in the form of their water-soluble salts, such as the alkali metal salts, preferably the sodium or potassium salts or the amine($NR_1R_2R_3$) salts, wherein

R_1, R_2 and R_3 are each independently of one another hydrogen, C_1-C_8alkyl, C_2-C_8alkenyl, C_1-C_8hydroxyalkyl, C_5-C_8cycloalkyl or polyalkenylenoxy-C_1-C_{18}alkyl, or

R_1, R_2 and R_3, together with the linking nitrogen atom, are unsubstituted or C_1-C_4alkyl-substituted morpholino.

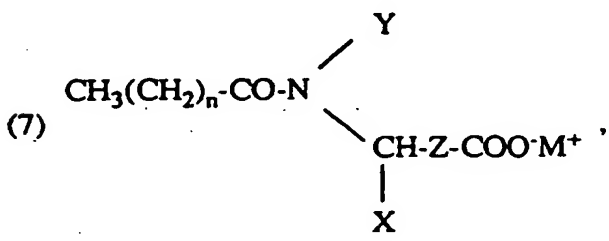
Component (b) can consist of only one compound of subclass (b_1) or also of mixtures of one or more than one compound of subclass (b_1), also together with components of further subclasses.

Preferably a combination of one or more than one compound of subclass (b_1) and one or more than one compound of subclass (b_2) is used.
Particularly preferred in this connection is a combination of cumene sulfonate and citric acid monohydrate.

Suitable components (c) are anionic, nonionic or zwitterionic and amphoteric synthetic, surface-active substances.

Suitable anionic surface-active substances are:

- sulfates, typically fatty alcohol sulfates, which contain 8 to 18 carbon atoms in the alkyl chain, e.g. sulfated lauryl alcohol;
- fatty alcohol ether sulfates, typically the acid esters or the salts thereof of a polyadduct of 2 to 30 mol of ethylene oxide with 1 mol of a C_8-C_{22}fatty alcohol;
- the alkali metal salts, ammonium salts or amine salts of C_8-C_{20}fatty acids, which are termed soaps, typically coconut fatty acid;
- alkylamide sulfates;
- alkylamine sulfates, typically monoethanolamine lauryl sulfate;
- alkylamide ether sulfates;
- alkylaryl polyether sulfates;
- monoglyceride sulfates;
- alkane sulfonates, containing 8 to 20 carbon atoms in the alkyl chain, e.g. dodecyl sulfonate;
- alkylamide sulfonates;
- alkylaryl sulfonates;
- α-olefin sulfonates;
- sulfosuccinic acid derivatives, typically alkyl sulfosuccinates, alkyl ether sulfosuccinates or alkyl sulfosuccinamide derivatives;
- N-[alkylamidoalkyl]amino acids of formula



(7)

wherein
X is hydrogen, C_1-C_4alkyl or $-COO^-M^+$,
Y is hydrogen or C_1-C_4alkyl,
Z is $-(CH_2)_{m_1-1}-$
m_1 is 1 to 5,
n_1 is an integer from 6 to 18, and
M is an alkali metal ion or an amine ion;
alkyl ether carboxylates and alkylaryl ether carboxylates of formula

(8) CH_3-X-Y-A ,

wherein

X is a radical $-(CH_2)_{5-19}O-$, $-(CH_2)_{5-11}-C_6H_4-O-$ or $-(CH_2)_{5-19}N(R)-$,

R is hydrogen or C_1-C_4alkyl,
Y is $-(CHCHO)_{1-50}-$,

A is $-(CH_2)_{m_2-1}COO^-M^+$ or $-P(=O)(O^-M^+)(O^-M^+)$

m_2 is 1 to 6, and
M is an alkali metal cation or an amine cation.

The anionic surfactants used may furthermore be fatty acid methyl taurides, alkylisothionates, fatty acid polypeptide condensates and fatty alcohol phosphoric acid esters. The alkyl radicals in these compounds preferably contain 8 to 24 carbon atoms.

The anionic surfactants are usually obtained in the form of their water-soluble salts, such as the alkali metal, ammonium or amine salts. Typical examples of such salts are lithium, sodium, potassium, ammonium, triethylamine, ethanolamine, diethanolamine or triethanolamine salts. It is preferred to use the sodium or potassium salts or the ammonium-($NR_1R_2R_3$) salts, wherein R_1, R_2 and R_3 are each independently of one another hydrogen, C_1-C_4alkyl or C_1-C_4hydroxyalkyl.

Very particularly preferred anionic surfactants in the novel formulation are

monoethanolamine lauryl sulfate or the alkali metal salts of fatty alcohol sulfates, preferably the sodium lauryl sulfate and the reaction product of 2 to 4 mol of ethylene oxide and sodium lauryl ether sulfate.

Suitable zwitterionic and amphoteric surfactants are C_8-C_{18}betaines, C_8-C_{18}sulfobetaines, C_8-C_{24}alkylamido-C_1-C_4alkylenebetaines, imidazoline carboxylates, alkylamphocarboxy carboxylic acids, alkylamphocarboxylic acids (e.g. lauroamphoglycinate) and N-alkyl-β-aminopropionates or N-alkyl-β-iminodipropionates. It is preferred to use the C_{10}-C_{20}alkylamido-C_1-C_4alkylenebetaines and, more particularly, cocoamidopropylbetaine.

Nonionic surfactants are typically derivatives of the adducts of propylene oxide/ethylene oxide having a molecular weight of 1000 to 15000, fatty alcohol ethoxylates (1-50 EO), alkylphenol polyglycol ethers (1-50 EO), ethoxylated carbohydrates, fatty acid glycol partial esters, typically diethylene glycol monostearate, fatty acid alkanolamides and fatty acid dialkanolamides, fatty acid alkanolamide ethoxylates and fatty acid amine oxides.

For component (c) may furthermore be used the salts of saturated and unsaturated C_8-C_{22}fatty acids, either by themselves, in admixture with each other or in admixture with the other surface-active substances cited for component (c). Illustrative examples of these fatty acids are typically capric, lauric, myristic, palmitic, stearic, arachic, behenic, dodecenoic, tetradecenoic, octadecenoic, oleic, eicosanic and erucic acid, as well as the technical mixtures of such acids, typically coconut fatty acid. These acids may be obtained in the form of salts, suitable cations being alkali metal cations such as sodium and potassium cations, metal atoms such as zinc atoms and aluminium atoms or nitrogen-containing organic compounds of sufficient alkalinity, typically amines or ethoxylated amines. These salt can also be prepared in situ.

Suitable components (d) are dihydric alcohols, preferably those containing 2 to 6 carbon atoms in the alkylene radical, typically ethylene glycol, 1,2- or 1,3-propanediol, 1,3-, 1,4- or 2,3-butanediol, 1,5-pentanediol and 1,6-hexanediol. 1,2-propanediol (propylene glycol) is preferred.

Component (e) is preferably ethanol, n-propanol and isopropanol, or a mixture of these alcohols.

Components (d) and (e) may also be obtained in admixture with each other.

The pH of the novel formulation is 3 to 10, preferably 3,5 to 5,5.

The novel formulations obtained as soap or syndet solutions may additionally comprise customary additives, typically sequestrants, dyes, perfume oils, thickeners or solidifiers (consistency regulators), emollients, UV absorbers, skin-protection agents, antioxidants, additives which improve the mechanical properties, such as dicarboxylic acids and/or Al, Zn, Ca, Mg salts of C_{14}-C_{22}fatty acids and, if desired, preservatives.

Soap formulations of the invention can be prepared by mixing components (a) and (b) and, optionally, (c), (d) and (e), in any order, with the requisite amount of water and stirring the mixture to homogeneity. The mixture is bulked to 100% with mains water or deinonised water. This procedure is a purely physical procedure. Accordingly, there is no chemical reaction of the individual components.

For disinfection and cleansing of the human skin and hands and of hard objects, the novel soap formulations can be applied thereto in dilute or undilute form, suitably in an amount of at least 2 ml, preferably in the undilute form, for hand disinfection.

The invention is illustrated by the following Examples. Parts and percentages are by weight.

Example 1:

0.075 % 2,4,4'-trichloro-2'-hydroxydiphenyl ether,
0.5 % monoethanolamine laurylsulfate
0.25 % sodium cumene sulfonate powder,
0.4 % citric acid monohydrate, and
0.5 % propylene glycol

are stirred to homogeneity and about 90% of the requisite water is then added. The pH is adjusted to 4.0 with monoethanolamine. Deionised water is then added to the solution to make up a total of 100 parts. The pH is checked again and, if necessary, monoethanolamine is added to adjust the pH to 4.0.

Example 2:

0.25 % 2,4,4'-trichloro-2'-hydroxydiphenyl ether

1.0 % monoethanolamine lauryl sulfate
2.5 % sodium cumene sulfonate powder,
1.5 % citric acid monohydrate, and
1.0 % propylene glycol
are stirred to homogeneity and about 90% of the requisite water is then added. The pH is adjusted to 4.0 with monoethanolamine. Deionised water is then added to the solution to make up a total of 100 parts. The pH is checked again and, if necessary, monoethanolamine is added to adjust the pH to 4.0.

Example 3:
0.15 % 2,4,4'-trichloro-2'-hydroxydiphenyl ether
1.0 % monoethanolamine lauryl sulfate
0.5 % sodium cumene sulfonate powder,
0.8 % citric acid monohydrate, and
5.0 % propylene glycol
are stirred to homogeneity and about 90% of the requisite water is then added. The pH is adjusted to 4.0 with monoethanolamine. Deionised water is then added to the solution to make up a total of 100 parts. The pH is checked again and, if necessary, monoethanolamine is added to adjust the pH to 4.0.

Example 4:
0.1 % 2,4,4'-trichloro-2'-hydroxydiphenyl ether
4.0 % monoethanolamine lauryl sulfate
0.5 % sodium cumene sulfonate powder, and
0.019 % NaOH
are stirred to homogeneity and about 90% of the requisite water is then added. The pH is adjusted to 9.1 with monoethanolamine. Deionised water is then added to the solution to make up a total of 100 parts. The pH is checked again and, if necessary, monoethanolamine is added to adjust the pH to 9.1.

Example 5: Test of the microbicidal activitiy of the novel formulations
The microbicidal activity (in decimal logarithms) of the novel formulations according to Examples 1 to 4 is determined with a suspension test. This test is used to assess the bactericidal activity of water-soluble antiseptics, disinfectants and of liquid soaps. The test consists in seeding the test product in selected dilutions with the test bacillus. After a certain contact time, aliquots is taken and the number of surviving bacilli is determined.

The difference between the number of the bacilli added and the number of the surviving bacilli is expressed as bacilli reduction in decimal logarithms. The concentration is 90%, the contact time is 30 seconds.

The following bacilli are used:

Example	1	2	3	4
Staph. aureus ATCC 9144	>5.0	>5.0	>5.0	>5.1
Strept. faecalis ATCC 10,541	>5.2	>5.2	>5.2	>5.2
E. Coli ATCC 10,536	4.3	5.1	4.0	*
P. aeruginosa CIP A-22	5.4	>5.4	>5.4	
Serratia marcescens ATCC 13,880	*	*	*	>5.4**

* not measured

** contact time 5 minutes

Values above 4 indicate good antimicrobial activity.

What is claimed is

1. A surface-active surfactant formulation, comprising

(a) 0.01 to 0.2% by weight of a microbicidal active substance,

(b) 0.1 to 7.5% by weight of one or more than one hydrotropic agent,

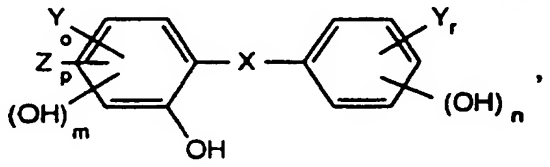
(c) 0 to 2% by weight of one or more than one synthetic surface-active substance or of a soap or of combinations of the cited substances and/or of a salt of a saturated and/or unsaturated C_8-C_{22}fatty acid,

(d) 0 to 10% by weight of a dihydric alcohol,

(e) 0 to 70% by weight of a monohydric alcohol, and

(f) mains water or deionised water to make up 100%.

2. A formulation according to claim 1, wherein component (a) is 2-hydroxydiphenyl ether, 2-hydroxydiphenylmethane and 2-hydroxydiphenyl thioether of the general formula

(1) 

,

wherein

X is oxygen, sulfur or -CH_2-,

Y is chloro or bromo,

Z is SO_2H, NO_2 or C_1-C_4alkyl,

r is 0 to 3,

o is 0 to 3,

p is 0 or 1,

m is 0 or 1, and

n is 0 or 1.

3. A formulation according to claim 2, wherein the compounds used for component (a) are those of formula (1), wherein

X is oxygen, sulfur or -CH_2-, and

Y is chloro or bromo,

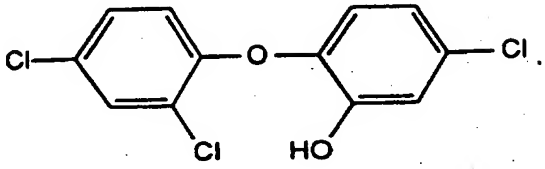
m is 0,

n is 0 or 1,

o is 1 or 2,
r is 1 or 2, and
p is 0.

4. A formulation according to claim 2 or 3, wherein the compounds used for component (a) are those of formula (1), wherein
X is oxygen, and
Y is chloro,
m is 0,
n is 0,
o is 1,
r is 2, and
p is 0.

5. A formulation according to claim 4, wherein the compound used for component (a) is that of formula

(2) 

6. A formulation according to any one of claims 1 to 5, wherein component (a) is used in an amount of 0.02 to 0.2% by weight.

7. A formulation according to any one of claims 1 to 6, wherein component (b_1) is a sulfonate, preferably a salt thereof of a terpenoid or of a mono- or binuclear aromatic compound.

8. A formulation according to claim 7, wherein the mono-or binuclear aromatic compound is a sulfonate of camphor, toluene, xylene, cumene or naphthene.

9. A formulation according to claim 7, wherein component (b) consists of only one compound of subclass (b_1) or also of a mixture of one or more than one compound of subclass (b_1), also with components of further subclasses.

10. A formulation according to any one of claims 1 to 7, wherein component (b) is a combination of one or more than one compound of subclass (b_1) and one or more than one compound of subclass (b_2).

11. A formulation according to claim 9, wherein a combination of cumene sulfonate and citric acid monohydrate is used.

12. A formulation of any one of claims 1 to 11, wherein the anionic surfactant is a fatty alcohol sulfate, which contains 8 to 18 carbon atoms in the chain.

13. A formulation according to claim 12, wherein the anionic surfactant is the alkali metal salt of the sulfated lauryl alcohol or of the monoethanolamine lauryl sulfate.

14. A formulation according to any one of claims 1 to 11, wherein component (b) is C_{10}-C_{20}alkylamido-C_1-C_4alkylenebetaine.

15. A formulation according to any one of claims 1 to 14, wherein the salt of a saturated and/or unsaturated C_8-C_{22}fatty acid corresponding to component (c) is selected from the group consisting of lauric, myristic, palmitic, stearic, arachic, behenic, dodecenoic, tetradecenoic, octadecenoic, oleic, eicosanic and erucic acid.

16. A formulation according to any one of claims 1 to 15, wherein component (d) is propylene glycol.

17. A formulation according to any one of claims 1 to 16, wherein component (e) is selected from the group consisting of ethanol, propanol, isopropanol and mixtures of these alcohols.

18. A formulation according to either claim 16 or claim 17, wherein components (d) and (e) are used in admixture with each other.

19. Use of an antimicrobial soap formulation according to any one of claims 1 to 18 for the disinfection and cleansing of the human skin and hands and of hard objects.

20. Use according to claim 19, wherein the antimicrobial soap formulation is in dilute or undilute form.

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(57) Abstract

The invention relates to surface-active soap formulations, comprising (a) 0.01 to 0.2 % by weight of a microbicidal active substance, (b) 0.1 to 7.5 % by weight of one or more than one hydrotropic agent, (c) 0 to 2 % by weight of one or more than one synthetic surface-active substance or of a soap or of combinations of the cited substances and/or of a salt of a saturated and/or unsaturated C_8-C_{22}fatty acid, (d) 0 to 10 % by weight of a dihydric alcohol, (e) 0 to 70 % by weight of a monohydric alcohol, and (f) mains water or deionised water to make up 100 %. The formulation is used for the disinfection and cleansing of the human skin and hands and of hard objects.

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B. FIELDS SEARCHED

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[X] Further documents are listed in the continuation of box C.

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